

DIRECT LIQUEFACTION: WHERE WE STAND
AND WHERE WE ARE SUPPOSED TO GO - AN OVERVIEW

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INTRODUCTION

The U.S. Department of Energy (DOE) precommercial activities in the 1970's and early 1980's established the commercial viability of direct coal liquefaction from a technical point of view. In the early 1980's, the direct liquefaction program was reoriented toward longer range process research and development activities with the objective of developing improved, lower cost technology. The DOE development program since then has made substantial and significant improvements in the process technology. Liquid yields have been improved by over 30 percent. On a balanced commercial plant basis, this translates to a production of about 3.3 barrels of liquids per ton of coal for the improved technology compared to 2.5 barrels for the technologies developed in the 1970's. The quality of the coal-derived distillates produced by the current technology has been substantially improved, but at the expenses of increased consumption of the upgrading catalyst. As a result, the currently produced liquids are of much higher quality than petroleum crudes in general. Other technology improvements include less severe operating conditions which favorably impact construction costs, operability and maintenance; and improved product slate flexibility, which allows greater freedom in producing products for specific market applications.

As a consequence of these improvements in technology, substantial cost reduction has been achieved. Current two-stage technology demonstrated at the Wilsonville, Alabama proof-of-concept (POC) facility gave estimated product costs at about \$38-40 per barrel. Because of improved quality, these products would be competitive with crude oil at about \$33-35 per barrel.

Supporting and exploratory research made critical contributions in some area of process development, e.g. by providing process analysis data leading to improved process operation and performance. F. Burke and coworkers at the Consolidation Coal R&D Division have done an excellent analytical and process evaluation work in this area. Similarly, D. Gray and the MITRE Corporation team provided valuable process economic information for comparative processes evaluation. To be noted, both activities are crosscutting in that they contribute to all DOE-supported processes under development. In addition, both supporting activities, through many years of experience and close cooperation between them and with the contractors working in process development, have reached a high degree of sophistication in various aspects of process evaluation so that, presently, they are capable of offering valuable research

guidance for the evaluation of novel research activities in coal liquefaction.

For more than a decade DOE has encouraged the generation of an extensive analytical and economic data bank on coal liquefaction processes and on related research activities, with the specific provision that the data and the ensuing process evaluation would be developed by competent sources independently from the actual process developers and from individual researchers.

Presently, we continue to pursue this endeavor by expanding the "teaming arrangement" promoted by Consolidation Coal and MITRE, and to support a similar "teaming" effort by A. Davis at Penn State University to establish an analytical data bank in support of the determination of the mechanisms and the kinetics of the rapid changes occurring in dispersed catalyst composition during the initial stages of liquefaction.

Consequently, we appear on the way to being prepared organizationally for the challenge presented to us in integrating into the current processing schemes the discoveries of present and future research and development studies.

PROGRAM OBJECTIVES/DIRECTION

In line with the current environmental and energy shortage concerns and with the effort to sustain coal as a viable option in competition with other energy sources, the main objective of the DOE's direct coal liquefaction current program is to develop the technology for the production of specification liquid fuels at a cost competitive with crude oil, within the next 5 to 7 years.

More specifically, the program calls for a proof-of concept demonstration, by 1997, of a system capable of producing liquid fuels from coal which complies with environmental requirements at cost of less than \$30 per barrel of oil equivalent and with lower greenhouse gas emissions than those from 1980 state-of-the-art coal liquefaction technology, or comparable to petroleum-based systems, and are suitable to be blended with petroleum stocks to produce and, perhaps, enhance the quality of reformulated gasoline, jet fuel and other high quality liquid transportation fuels.

The key issue is the integration of coal-derived liquids into existing petroleum refining systems which would require no major modifications to the fuel distribution and storage infrastructure which currently exists.

In the evaluation and planning of present and future research and development activities, we need to provide answers to two important questions: How should the present activities in the fundamental and applied research at the university and national laboratory level be changed in order that the remaining scientific and technical challenges can be addressed? And, what are the most important feedback information that the process developers and process evaluation teams can provide to the scientific community in order to evidence the most compelling research and development needs to

overcome the current challenges and provide a better understanding of the reactions involved in coal liquefaction?

The first step is the recognition that all of the knowledge in this field does not and cannot reside in a single discipline. We need to support in a substantive way a policy of multi-disciplinary research teamwork and to further encourage cooperative efforts between the fundamental and applied research and bench scale development of integrated process activities.

The excellent study done by a panel group on research needs assessment for coal liquefaction (COLIRN), headed by H.D. Schindler, and sponsored by the DOE Office of Energy Research, Office of Program Analysis, resulted in a publication which provided important recommendations to be implemented by the fundamental and applied researchers in this field. The recommendations were reviewed and approved by the representatives of DOE Office of Fossil Energy, with some reservations about the usefulness of chemical pretreatments in the overall liquefaction processing scheme and the use of novel catalysts prior to gain a better understanding of the mechanism and the kinetics involved in the initial stages of coal liquefaction.

We agree with the conclusion of the COLIRN panel which has challenged the conventional view of the chemistry and the mechanisms of direct liquefaction--that the initial reactions of liquefaction involve thermal homolytic bond cleavage with stabilization of the free radicals formed-- and is being replaced by the recent work on a deeper understanding of bond-breaking and bond-forming processes via hydrogen transfer. This new model of coal depolymerization may explain the cleavage of bonds which are too strong to be broken thermally at liquefaction conditions.

The COLIRN panel agreed that these new understandings of structure and mechanisms are expected to lead to more efficient liquefaction processes, which, eventually, need to be developed before coal liquefaction can be commercialized.

In particular, the panel placed the highest priority on identifying the coal structures responsible for retrograde reactions and the reaction mechanisms and kinetics of these reactions. The panel also placed high priority on developing a coal structure-reactivity relationship, develop kinetic models of liquefaction, and develop intrinsic quantitative rate expressions as a basis for understanding initial reaction paths during coal dissolution.

Among the remaining recommendations the panel scored lower priority for the study on catalytic hydrogenation and cracking functions mechanism to establish their interaction and to determine the effects of thermal reaction on these functions. However, current data from the Wilsonville POC unit indicate that the thermal/catalytic staged liquefaction mode gives substantially higher coal conversion than the catalytic/thermal mode, and this fact is mistifying process evaluators and, as a consequence, a better understanding of the contribution and of the reaction

mechanisms attributed to the thermal and catalytic functions would certainly require a much higher research priority because it is quite evident that our knowledge in this important area is indeed insufficient.

CONCLUSIVE REMARKS

The current advanced research program needs to be reoriented to follow more closely the recommendations of the COLIRN panel and the direction suggested by the process development program, which require a clearer understanding of the initial reaction paths, particularly the reactions involved in the preconversion step and those of the attendant catalyst deactivation mechanism. The acquisition of this knowledge should lead to substantially more efficient liquefaction processes.

According to J. Larsen³ there are three areas which are crucial to our state of knowledge in coal processing: coal structure, coal as reaction environment, and which reactions are actually occurring in coals.

We agree with J. Larsen that our knowledge in all three areas is sufficiently uncertain to make extrapolation from routine model compound studies to coals quite unreliable. And we need to add that similar extrapolation unreliabilities exist for the current catalyst studies and applications, solvent extraction, alkylation, pyrolysis, chemical pretreatment, water supported reactions and many other approaches fashionable in past and present advanced research activities because they suffer from the same lack of fundamental knowledge of coal structure, reaction mechanisms and kinetics to make them viable for further scaleup consideration.

The process development activities is also at crucial turning point because we are finding that coal derived materials, such as resids and the quality of the solvent, which are used in the recycle stream, exhibit larger beneficial effects in coal liquefaction than the comparison of novel catalyst formulation versus existing commercial catalysts.

Consequently, the commitment of DOE to develop the technology for the production of specification liquid fuels from coal, at a cost competitive with crude oil, within the next 5 to 7 years, will require a reorientation of the advanced research program, increases in both scope and pace of the fundamental and applied research effort, perhaps by teaming arrangements with the process development and process evaluation programs, to produce a timely resolution of the technical issues in order to meet the requirements dictated by the current and perhaps more restrictive future environmental and economic constraints for coal derived liquid fuels.

If a strong and clear message should be given to the participants of this "Symposium on Coal Reaction Mechanism", from the content of this brief overview, it would sound like a mimic of the famous message contained in the inaugural address of the late president of the U.S., J.F. Kennedy:

"Ask not what you can do for coal processing",
"Ask, first, what coal structure/reactions can do for you".

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